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Title	Squeezed spin states
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Citation	Physical Review A. 1993, 47(6), p. 5138-5143
Version Type	VoR
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Squeezed spin states

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(Received 12 February 1991; revised manuscript received 3 December 1992)

The basic concept of *squeezed spin states* is established and the principles for their generation are discussed. Two proposed mechanisms, referred to as *one-axis twisting* and *two-axis countertwisting*, are shown to reduce the standard quantum noise $S/2$ of the coherent S-spin state down to $\frac{1}{2}(S/3)^{1/3}$ and $\frac{1}{2}$, respectively. Implementations of spin squeezing in interferometers are also discussed.

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I. INTRODUCTION

Squeezing, which redistributes quantum fluctuations between two noncommuting observables while preserving the minimum uncertainty product, has been extensively studied in boson systems [1]. A radiation field is said to be squeezed if the uncertainty of one quadrature amplitude $\langle \Delta a_i{}^2 \rangle$ is smaller than the standard quantum limit (SQL) of $\frac{1}{4}$. Quantum-mechanical correlations between photons established through nonlinear interactions play an essential role in the generation of squeezed states of light.

Spin or angular momentum systems [2] have often been regarded as squeezed if the uncertainty of one spin component, say $\langle \Delta S_x{}^2 \rangle$ or $\langle \Delta S_y{}^2 \rangle$, is smaller than $\frac{1}{2}|\langle S_z \rangle|$ [3]. This definition implies that a *coherent spin state* (CSS) [4] *is* already squeezed if it is placed in an appropriate system of coordinates, and also that spin can be squeezed by just rotating the CSS. Squeezed light emission from an atomic system in a certain CSS [5] has been regarded as evidence justifying this definition [3]. However, it is by no means obvious that we can judge the squeezing of spin by referring to the uncertainty of another (i.e., photon) system interacting with it. Moreover, the definition itself is problematic because it does not reflect quantum correlations but depends solely on the particular coordinate system. The reduced variance of a spin component does not necessarily mean squeezing in spin systems. The squeezing of spin is not as straightforward as the squeezing of bosons since their uncertainty relationships are essentially different [6].

The previous definition of squeezing in a spin system is also inappropriate from a practical viewpoint since it fails to correctly locate the SQL that is to be overcome by squeezing. It is known that an interferometer can be described as a spin system [7], yet the SQL of the interferometric phase sensitivity can never be overcome by a mere rotation of the CSS [8]. A spin state that improves the interferometric phase sensitivity beyond the SQL has been mathematically constructed [7, 8], and it is different from the CSS. These facts raise serious questions about whether CSS's are really qualified as squeezed states or whether there are other states that are more qualified to be called *squeezed spin states* (SSS's). Also, a problem remains about how such states, if they exist, can be generated from experimentally available CSS's.

How to define and achieve spin squeezing is an important problem since spin can describe such diverse physical systems as the real spin of particles and magnons, collective two-level atoms [9], Cooper pairs in superconductors, and macroscopic two-state systems [10] like interferometers [7, 8], and Josephson junctions.

This paper establishes the concept of *squeezed spin states* (SSS's) and discusses the general principles for generating them. This paper is organized as follows. Section II describes the basic concept of spin squeezing in terms of quantum correlations among elementary spins. Section III proposes two mechanisms for spin squeezing and discusses their limits in quantum noise reduction. First, we propose a *one-axis twisting* mechanism as a building block for spin squeezing. Then, a *two-axis countertwisting* mechanism is introduced as a natural extension of one-axis twisting; this is shown to further reduce the quantum noise. Section IV discusses implementations of the twisting mechanisms in interferometers and two-level atoms. The Appendix derives some formulas that are necessary for calculating the moments.

II. BASIC CONCEPT OF SQUEEZING IN SPIN SYSTEMS

The spin or angular momentum system $\mathbf{S} = (S_x, S_y, S_z)$ is governed by the cyclic commutation relations, $[S_i, S_j] = i\epsilon_{ijk}S_k$, where suffixes i, j, k denote the components in any three orthogonal directions and ϵ_{ijk} is the Levi-Civita symbol. The associated uncertainty relationship is $\langle \Delta S_i{}^2 \rangle \langle \Delta S_j{}^2 \rangle \geq \frac{1}{4}|\langle S_k \rangle|^2$. A CSS $|\theta, \phi\rangle$ is defined as an eigenstate of a spin component in the (θ, ϕ) direction, $S_{\theta,\phi} = S_x \sin\theta\cos\phi + S_y \sin\theta\sin\phi + S_z\cos\theta$, with eigenvalue S, where θ and ϕ denote the polar and azimuth angles. The CSS satisfies the minimum uncertainty relationship with uncertainties $\frac{S}{2}$ equally distributed over any two orthogonal components normal to the (θ, ϕ) direction. Therefore, the spin vector $\mathbf{S}$ in a CSS can be conceived as a cone [11] as schematically shown in Fig. 1(a). In more rigorous words, the CSS has an isotropic quasiprobability distribution (QPD) [12] in a spherical phase space as shown in Figs. 2(a) and 3(a).

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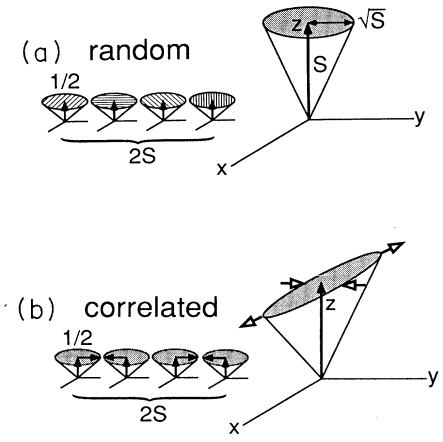

FIG. 1. Schematic illustrations of S-spin states in terms of $2S$ individual 1/2 spins. (a) Coherent spin state constructed from $2S$ uncorrelated 1/2 spins. (b) Squeezed spin state constructed from $2S$ correlated 1/2 spins.

An S-spin system can be regarded as a collective system consisting of $2S$ elementary $\frac{1}{2}$ spins [13]. Any pure state of a $\frac{1}{2}$ spin is a CSS $|\theta_k,\phi_k\rangle_k = \cos\frac{\theta_k}{2}|\uparrow\rangle_k + e^{i\phi_k}\sin\frac{\theta_k}{2}|\downarrow\rangle_k$, where $|\uparrow\rangle_k$ ($|\downarrow\rangle_k$) is the eigenstate of S_z with the eigenvalue $\frac{1}{2}$ ($-\frac{1}{2}$) in the kth $\frac{1}{2}$-spin system. The components normal to (θ_k,ϕ_k) are completely uncertain, having a variance of $\frac{1}{4}$. The S-spin CSS $|\theta,\phi\rangle$ is equivalent to a set of $2S$ elementary spins all pointing in the same mean direction (θ,ϕ) as shown in Fig. 1(a). Since there are no quantum-mechanical correlations among these elementary spins, the variance of the components normal to the mean direction is simply the sum of the variances of the individual elementary spins, and is thus $\frac{S}{2}$. Now, if appropriate quantum-mechanical correlations are established among the elementary spins as schematically shown in Fig. 1(b), it is possible to partly cancel out fluctuations in one direction at the expense of those enhanced in the other direction. This is the basic concept of *spin squeezing*. The spin vector $\mathbf{S}$ in a squeezed spin state can be conceived as an elliptical cone [7] as schematically shown in Fig. 1(b). Such a state has an elliptical QPD in contrast to the isotropic one for a CSS. We regard spin as *squeezed* only if the variance of one spin component normal to the mean spin vector is smaller than the SQL of $\frac{S}{2}$. We have thus excluded mere mathematical coordinate dependency and included quantum correlation in our notion of squeezing. This notion naturally reflects the improved performance of spin systems.

To correlate the elementary spins requires a nonlinear interaction because a linear Hamiltonian merely rotates the individual spins and does not establish quantum correlations among them [14]. A $\frac{1}{2}$-spin system cannot be squeezed since it is equivalent to a system with only one elementary spin, which therefore has no partner to be correlated with.

III. SPIN SQUEEZING BY NONLINEAR INTERACTIONS

A. One-axis twisting

Now we demonstrate how the spin can be squeezed by nonlinear interaction. We consider a class of unitary transformations $U(t) = \exp[-itF(S_z)]$ generated by the Hamiltonian $H = \hbar F(S_z)$ and see how they deform the noise distribution. The ladder operators $S_\pm \equiv S_x \pm iS_y$ evolve as

$$S_+(t) = U^\dagger S_+(0)U = S_+(0)\exp[itf(S_z)]$$

and $S_-(t) = [S_+(t)]^\dagger$, where

$$f(S_z) = F(S_z+1) - F(S_z).$$

The lowest-order nonlinear interaction $F(S_z) = \chi S_z^2$ leads to $f(S_z) = 2\chi(S_z + \frac{1}{2})$, rotation proportional to S_z, which *twists* the quantum fluctuations as shown in Fig. 2(b) [15]. This is analogous to *self-phase modulation* in the photon system [16]. The components after twisting are given by $\tilde{S}_x = \frac{1}{2}[S_+e^{i\mu(S_z+1/2)} + e^{-i\mu(S_z+1/2)}S_-]$ and $\tilde{S}_y = \frac{1}{2i}[S_+e^{i\mu(S_z+1/2)} - e^{-i\mu(S_z+1/2)}S_-]$, where $\tilde{S}_i$ and S_i denote $S_i(t)$ and $S_i(0)$, and $\mu \equiv 2\chi t$. With the CSS $|\frac{\pi}{2},0\rangle = 2^{-S}\sum_{k=0}^{2S}\binom{2S}{k}^{1/2}|S,S-k\rangle$ as an initial state, we show in Figs. 2(b) and 2(c) how uncertainties are deformed by *twisting* as μ increases. Uncertainties are redistributed between certain orthogonal components in the y-z plane.

Let us rotate the distribution around the x axis by the unitary transformation $\bar{\mathbf{S}} = \exp(i\nu\tilde{S}_x)\tilde{\mathbf{S}}\exp(-i\nu\tilde{S}_x)$ to see how the uncertainties are redistributed. The means and variances become

$$\langle\bar{S}_x\rangle = S\cos^{2S-1}\tfrac{\mu}{2}, \quad \langle\bar{S}_y\rangle = 0, \quad \langle\bar{S}_z\rangle = 0, \tag{1}$$

$$\langle\Delta\bar{S}_x^2\rangle = \tfrac{S}{2}[2S(1-\cos^{2(2S-1)}\tfrac{\mu}{2}) - (S-\tfrac{1}{2})A], \tag{2}$$

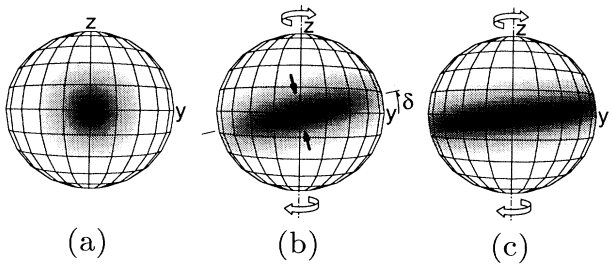

FIG. 2. State evolutions by one-axis twisting in terms of the quasiprobability distribution (QPD) on the sphere for $S = 20$. The densities of the figures are normalized by the maximum value $Q_{\max}$ of $Q(\theta,\phi)$. (a) shows the initial coherent spin state $|\theta{=}\frac{\pi}{2},\phi{=}0\rangle$ ($Q_{\max} = 1$). (b) and (c) show one-axis twisted states generated by the unitary transformation $U = \exp[-i\mu S_z^2/2]$; (b) optimally squeezed at $\mu = 0.199$ ($Q_{\max} = 0.445$) and (c) excessively twisted at $\mu = 0.399$ ($Q_{\max} = 0.241$). Although not clear from the figure, the QPD of (c) deviates from a geodesic (swirliness).

$$\langle \Delta \bar{S}_z^2 \rangle = \frac{S}{2}\{1 + \tfrac{1}{2}(S - \tfrac{1}{2})[A \pm \sqrt{A^2 + B^2}\cos(2\nu + 2\delta)]\}, \tag{3}$$

where we define $A = 1 - \cos^{2S-2}\mu$, $B = 4\sin\frac{\mu}{2}\cos^{2S-2}\frac{\mu}{2}$, and $\delta = \frac{1}{2}\arctan\frac{B}{A}$. Equation (3) shows the anisotropically distributed quantum fluctuations of the SSS. Our primary concern is to minimize one of the variances in Eq. (3). The term $\langle \Delta \bar{S}_y^2 \rangle$ is minimized and $\langle \Delta \bar{S}_z^2 \rangle$ is maximized when $\nu = \frac{\pi}{2} - \delta$. Likewise $\langle \Delta \bar{S}_z^2 \rangle$ is minimized and $\langle \Delta \bar{S}_y^2 \rangle$ is maximized when $\nu = -\delta$. The increased (upper sign) and decreased (lower sign) variances are written as

$$V_\pm = \frac{S}{2}\{[1 + \tfrac{1}{2}(S - \tfrac{1}{2})A] \pm \tfrac{1}{2}(S - \tfrac{1}{2})\sqrt{A^2 + B^2}]\}. \tag{4}$$

For $S \gg 1$ and $|\mu| \ll 1$, they can be approximated as

$$V_+ \approx \frac{S}{2}4\alpha^2, \quad V_- \approx \frac{S}{2}\left(\frac{1}{4\alpha^2} + \frac{2}{3}\beta^2\right), \tag{5}$$

where we have set $\alpha = \frac{1}{2}S\mu$ and $\beta = \frac{1}{4}S\mu^2$ and assumed $|\alpha| > 1$ and $\beta \ll 1$, but kept the terms up to the lowest order in β to take into account the spherical nature of the phase space and the *swirliness* of the QPD — the deviations from a geodesic. Also we get $\delta \approx \frac{1}{2}\arctan(1/\alpha)$, $\langle \bar{S}_x \rangle \approx S(1 - \beta)$, and $\langle \Delta \bar{S}_x{}^2 \rangle \approx 2\alpha^2$. The uncertainty $\sqrt{V_-}$ is suppressed by a factor of $2|\alpha|$ compared to that of the initial CSS, while $\sqrt{V_+}$ is enhanced by the same factor. The reduced variance V_- reaches its minimum $V_{\min} \approx \frac{1}{2}(\frac{S}{3})^{1/3}$ at $|\mu| = \mu_0 = 24^{1/6}S^{-2/3}$ when the second term on the right-hand side of Eq. (5) becomes comparable to the first term. The exact minimum attainable variances obtained from Eq. (4) are plotted as a function of S in Fig. 4. They are even smaller than the above approximate expression for small S and asymptotically approach it for large S. The normalized uncertainty product $U_{yz} \equiv 4\langle \Delta S_y{}^2 \rangle \langle \Delta S_z{}^2 \rangle / |\langle S_x \rangle|^2$ is calculated to be $U_{yz} \approx 1 + (\frac{\mu}{\mu_0})^6$; therefore, the state remains almost in the minimum uncertainty state for $|\mu| < \mu_0$. Figure 2(b) shows the QPD of the optimally squeezed state which gives the minimum variance for $S = 20$ and Fig. 2(c) shows that of an excessively twisted state. The minimum attainable uncertainty of one-axis twisting is limited by the swirliness of the QPD.

B. Two-axis countertwisting

The swirliness cancels out if the QPD is simultaneously twisted clockwise and counterclockwise about two orthogonal axes, both normal to the mean spin vector of the initial CSS as shown in Fig. 3(b). Let us squeeze the initial CSS $|0, \phi\rangle$ with respect to S_x and S_y by twisting it about the two axes in the $\theta = \frac{\pi}{2}, \phi = \pm\frac{\pi}{4}$ directions. The Hamiltonian of *two-axis countertwisting* is written as

$$H = \hbar\chi(S^2_{\frac{\pi}{2},\frac{\pi}{4}} - S^2_{\frac{\pi}{2},-\frac{\pi}{4}}) = \frac{\hbar\chi}{2i}(S_+^2 - S_-^2). \tag{6}$$

The minimum attainable variance of the two-axis SSS is less than $\frac{1}{2}$ for small S and asymptotically approaches $\frac{1}{2}$ as S increases, as shown in Fig. 4. The QPD shown in Fig. 3(b) is that of the optimally squeezed state that gives the minimum variance for $S = 20$. The QPD is shrunk along a geodesic on the sphere and stretched along the orthogonal geodesic. When the QPD spans almost half of the sphere, the reduced variance attains a minimum of $\frac{1}{2}$ while the enhanced one reaches $\frac{S^2}{2}$. As the length of the mean spin vector remains on the order of S, the state is still close to the minimum uncertainty state. If $\mu = 4\chi t$ exceeds the optimal value, the QPD splits into two parts as shown in Fig. 3(c).

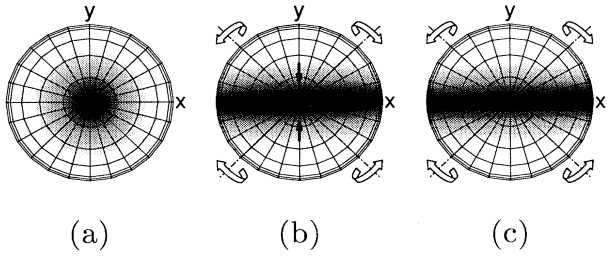

FIG. 3. State evolutions by two-axis countertwisting in terms of the quasiprobability distribution (QPD) on the sphere for $S = 20$. The densities of the figures are normalized by the maximum value $Q_{\max}$ of $Q(\theta, \phi)$. (a) shows the initial coherent spin state $|\theta{=}0, \phi\rangle$ ($Q_{\max} = 1$). (b) and (c) are two-axis countertwisted states generated by the unitary transformation $U = \exp[-i\mu(S^2_{\frac{\pi}{2},\frac{\pi}{4}} - S^2_{\frac{\pi}{2},-\frac{\pi}{4}})/4]$; (b) optimally squeezed at $\mu = 0.203$ ($Q_{\max} = 0.252$) and (c) excessively twisted at $\mu = 0.248$ ($Q_{\max} = 0.187$), where the QPD splits into two parts.

In spin systems, the squeezing occurs on the phase sphere (spherical phase space). Unlike boson squeezing, the QPD cannot be homogeneously or globally squeezed in one direction over the whole phase space. If a spin

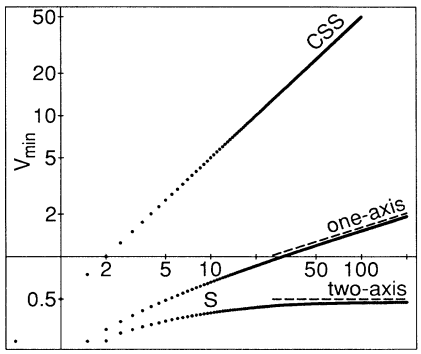

FIG. 4. The minimum attainable variances of the spin component normal to the mean spin vector as a function of S for one-axis squeezed spin states and two-axis squeezed spin states. They asymptotically approach $\frac{1}{2}(\frac{S}{3})^{1/3}$ and $\frac{1}{2}$ (broken lines) for increasing S. The special case of $S = 1$ where they reach 0 is not shown. The variances of coherent spin states, $\frac{S}{2}$, are also plotted for comparison.

component is shrunk around a certain point on the sphere, it must be stretched around another point. This imposes a fundamental restriction on the reduction in quantum noise. The two-axis countertwisting mechanism is the one that achieves the maximal noise reduction. The $S = 1$ spin system is an exception where both one-axis and two-axis twisting can completely squeeze out the uncertainty of one component and generate the eigenstate of that component with the eigenvalue 0.

IV. IMPLEMENTATIONS

Finally, we briefly discuss implementations of our twisting Hamiltonians in two-state systems.

A. Interferometers

Let us consider N quanta partitioned by a 50% beamsplitter into two outputs, A and B. Each quantum is in a superposition of the state in which the particle exists in A but not in B and the state in which it exists in B but not in A, as shown in Fig. 5. The former (latter) is an eigenstate of $\sigma_k^z = \frac{1}{2}(a_k^\dagger a_k - b_k^\dagger b_k)$ with eigenvalue $\pm\frac{1}{2}$ which corresponds to $|\uparrow\rangle$ ($|\downarrow\rangle$) of $\frac{1}{2}$ spin. Beamsplitters and phase shifters rotate this abstract "*spin.*"

At the input port of the beamsplitter, N particles all exist in A, namely, all N of the $\frac{1}{2}$ spins are in $|\uparrow\rangle$. This is a CSS $|\theta = 0, \phi\rangle$ of $\frac{N}{2}$ spin. It is also an eigenstate of $\mathbf{S}^2$ with eigenvalue $\frac{N}{2}(\frac{N}{2}+1)$ and continues to be so as long as the Hamiltonian commutes with $\mathbf{S}^2$. For example, dispersion-less beamsplitters and phase shifters rotate all N spins in the same manner and preserve $\mathbf{S}^2$. Under these conditions, the system continues to behave as a spin $\frac{N}{2}$.

The mathematical relationships between the partitioned N quanta and spin $\frac{N}{2}$ are [7, 8]

$$S_+ \equiv \sum_k a_k^\dagger b_k = S_x + iS_y, \qquad (7)$$

$$S_z \equiv \tfrac{1}{2}(N_A - N_B) \quad \left(N_A \equiv \sum_k a_k^\dagger a_k,\ N_B \equiv \sum_k b_k^\dagger b_k\right).$$

The creation and annihilation operators $a_k^\dagger$ and a_k express the quasimonochromatic field of arm A, and $b_k^\dagger$ and b_k those of B, which obey either boson commutation relations or fermion anticommutation relations. The operator S_+ coherently (without changing mode index k) transfers a particle from B to A and its phase represents the relative phase of A and B. The vertical component S_z represents half of the particle number difference between A and B.

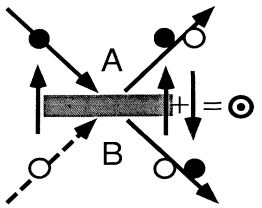

FIG. 5. A particle in an interferometer as an abstract spin 1/2. Filled circles mean that a particle exists and open circles mean that it does not. The arrow indicating which arm the particle exists in behaves like a spin 1/2.

How can we twist this spin $\frac{N}{2}$ system? Physically, we can twist the spin about the z axis by modulating the relative phase of A and B by the population difference between A and B.

For example, if the phase of A varies in proportion to N_A and that of B in proportion to N_B, then the relative phase is modulated by the population difference. For photons, this can be achieved by inserting an optical Kerr medium into each arm and turning on the self-phase modulations [16]. The interaction Hamiltonian is given by $H_I = \hbar\chi(N_A^2 + N_B^2) = 2\hbar\chi(N^2/4 + S_z^2)$, which performs one-axis twisting since N is a constant of motion.

Alternatively, if the phase of A varies in proportion to N_B and that of B in proportion to N_A, the result is also modulation of the relative phase by the population difference, but the sign is opposite. For charged particles, this can be achieved as a mutual phase modulation due to Coulomb interaction between particles of each arm [17]. The interaction Hamiltonian is given by $H_I = \hbar\chi N_A N_B = \hbar\chi(N^2/4 - S_z^2)$, which performs one-axis twisting.

The two-axis countertwisting Hamiltonian in Eq. (6) suggests an interaction that coherently transfers two particles at the same time. This might be achieved by four-wave mixing [7].

What is the merit of squeezing this spin $\frac{N}{2}$ system? When N quanta are equally partitioned, i.e., $\langle S_z\rangle = 0$, the uncertainties of particle-number difference and phase difference are given by

$$\delta N \equiv \langle[\Delta(N_A - N_B)]^2\rangle = 2\langle\Delta S_z^2\rangle^{1/2}, \qquad (8)$$

$$\delta\phi \equiv \langle[\Delta(\phi_A - \phi_B)]^2\rangle \approx \langle\Delta S_y^2\rangle^{1/2}/|\langle S_x\rangle|, \qquad (9)$$

assuming that the mean spin vector $\langle\mathbf{S}\rangle$ is parallel to the x axis [17]. The uncertainty relationship $\delta N\delta\phi \geq 1$ follows from the spin uncertainty relationships. A phase difference smaller than $\delta\phi$ is not detectable by interferometric measurement. For an ordinary 50% linear beamsplitter, $\delta N = \sqrt{N}$ and $\delta\phi \approx 1/\sqrt{N}$ because the outputs form an CSS $|\frac{\pi}{2}, 0\rangle$. Spin squeezing can reduce $\delta\phi$ without violating the uncertainty relationship and therefore it improves the interferometric sensitivity.

This is a totally new possibility for fermions since no fermion analog of boson squeezed state [18] has been found [19]. The application of spin squeezing in partition and interferometry will be discussed in detail elsewhere [20].

B. Two-level atoms

A collection of N two-level atoms can be described as a spin system of $S = \frac{N}{2}$ [9]. Correspondence is basically the same as above by replacing the arm A with the upper state $|A\rangle$, B with the lower state $|B\rangle$, and the kth mode with the kth atom. Each atom is represented by an ab-

stract spin $\frac{1}{2}$. The populations of the upper and lower states are N_A and N_B, respectively. The vertical component S_z corresponds to half the population difference. The horizontal components S_x and S_y represent the two quadrature-phase amplitudes of the dipole moment.

If all of the atoms are in the upper (lower) state, the collective system is in a CSS $|\theta = 0, \phi\rangle$ ($|\theta = \pi, \phi\rangle$). The dipole interaction of the collective atomic system with classical electromagnetic wave rotates the spin vector [4], but does not offer spin squeezing.

The one-axis twisting Hamiltonian corresponds to the energy propotional to the square of the population difference. The two-axis twisting Hamiltonian corresponds to the simultaneous excitation-deexcitation of two atoms. Although realistic physical schemes are yet to be found, these nonlinear Hamiltonians will provide some clues in the search for squeezed atomic states [21].

V. CONCLUSIONS

In conclusion, we have clarified the concept of squeezing in spin systems. An S-spin system is squeezed only if one of the components normal to the mean spin vector has a variance smaller than $S/2$. We have shown the principle for spin squeezing. The spin can be squeezed by establishing quantum correlations among elementary spins. We have proposed two fundamental mechanisms for spin squeezing, and discussed their limits of noise reduction. One-axis twisting can reduce the noise down to the order of $S^{1/3}$ and two-axis countertwisting can reduce it to 1/2. We have also discussed implementations of spin squeezing in interferometers.

APPENDIX: CALCULATIONS OF TWISTED MOMENT

In this appendix, we derive some formulas for calculating the moments of twisted spin operators. A coherent spin state $|\theta, \phi\rangle$ can be expanded in terms of S_z eigenstates $|S, k\rangle$ as follows [4]:

$$|\theta, \phi\rangle = \left(1 + \tan^2 \frac{\theta}{2}\right)^{-S} \times \sum_{k=0}^{2S} \left(e^{i\phi} \tan \frac{\theta}{2}\right)^k \binom{2S}{k}^{1/2} |S, S-k\rangle.$$

In the following, we assume that the initial state is a CSS:

$$\left|\frac{\pi}{2}, 0\right\rangle = 2^{-S} \sum_{k=0}^{2S} \binom{2S}{k}^{1/2} |S, S-k\rangle,$$

and write the expectation value of an operator O with respect to this state simply as $\langle O\rangle$.

The first moments of the spin components are calculated from

$$\langle S_+ \exp[i\mu(S_z+\tfrac{1}{2})]\rangle = 2^{-2S} \sum_{k=0}^{2S} \sum_{l=0}^{2S} \binom{2S}{k}^{1/2} \binom{2S}{l}^{1/2} \langle S, S-k|S_+ \exp[i\mu(S_z+\tfrac{1}{2})]|S, S-l\rangle = S\left(\cos \frac{\mu}{2}\right)^{2S-1} \quad \text{(A1)}$$

and its complex conjugate.

The calculation of the second moments requires the following two formulas. In a similar manner as Eq. (A1), we obtain

$$\langle \{S_+ \exp[i\mu(S_z+\tfrac{1}{2})]\}^2\rangle = S(S-\tfrac{1}{2})(\cos\mu)^{2S-2}. \quad \text{(A2)}$$

Differentiating Eq. (A1) with respect to μ yields

$$\langle iS_+ \exp[i\mu(S_z+\tfrac{1}{2})](S_z+\tfrac{1}{2})\rangle = -S(S-\tfrac{1}{2})\left(\cos \frac{\mu}{2}\right)^{2S-2} \sin \frac{\mu}{2}. \quad \text{(A3)}$$

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